

## Response to Anonymous Referee #2

Reviewer comments are in black, responses are in blue

### General Comments:

The authors examine and discuss factors important for fast, stable spinning of spherical rotors for magic angle spinning solid-state NMR experiments. They show that a stator for spherical rotors can be integrated into a commercial NMR probe head in a straightforward manner. They present measurements of spinning speeds and spinning stability achieved with eight different turbine groove designs, including one design with no grooves. The spinning rate achieved at a given pressure is shown to be sensitive to the surface grooves, with moderate spinning rate improvements observed for some groove geometries over others, and over a smooth rotor. Finally, they present a theoretical motivation for the spinning stability they observed in spherical rotors based on an analysis of the principal values of a spherical rotor's moment of inertia tensor.

The results presented here are broadly interesting to the solid-state NMR community. Spherical rotors represent an alternative to the conventionally used cylindrical rotors, with several potential benefits which the authors outline. While much development work is still necessary, this work reports progress which is likely to spur further investigation into spherical rotors, and into other alternative rotor designs.

### Specific Comments:

It would be interesting to hear comments about how spherical rotor stability is influenced by the sample, beyond what was included at the end of the manuscript.

To address this concern, we have added as supplementary material an interactive Mathematica document which allows the reader to independently adjust the densities for the sample, caps, and rotor in order to see the effect on the moments of inertia as a function of normalized inner radius. We have added additional discussion on this topic to this document.

NMR data acquired on a sample spun in rotor H were acquired with a 3.5 kHz spinning speed, which is below the maximum spinning speed reported for rotor H in Figure 3a. Was 3.5 kHz chosen for stability reasons?

As shown in Figure 3A, rotor H is capable of spinning stably at 4 kHz at 4 bar of air pressure. However, we exercised reasonable caution for the NMR experiment, as 3.5 kHz can be achieved with nearly half the air pressure required to reach 4 kHz, and the experimental results achieve the same goal.

Was any consideration given when designing deep turbine grooves to how these grooves might alter the moment of inertia tensor of the spherical rotors?

At the time of their design, we were not considering that removing material might have a significant effect on the inertia tensor of the rotor itself, and instead assumed that the grooves would be a negligible change. However, it is possible that removing a significant amount of material from the outer equatorial region could have had an impact on their stability by changing the inertia tensor. We still maintain however, that the primary effect causing rotors E, F, and G to not spin stably was due to the excess space allowing for complex and turbulent fluid flows in the stator cup. The motion of rotors E, F and G in the

stator is random and chaotic even at very low pressures/flow rates, while for the other rotors, they simply don't spin until a certain pressure threshold is reached and then beyond that, spin up smoothly. More research is needed to explore the fluid dynamics of the gas flow within the stator cup, as even for the simplest case (Rotor H) we expect the fluid flow to be very complex and require a complete 3D description. For now, the guidance on turbine groove development is to make them as shallow as possible, which should also minimize changes to the spherical ring inertia tensor.

Technical corrections:

The formatting of several of the references is somewhat strange, with the doi appearing twice in multiple references.

We have now corrected this issue.